Enhanced Magnetoresistance Induced by Spin Transfer Torques in Granular Films with a Magnetic Field

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Abstract

Spin-transfer torques (STT) provide a mechanism to alter the magnetic configurations of magnetic heterostructures, a result previously only achieved by an external magnetic field. In granular solids, we demonstrate a new form of STT effect that can be exploited to induce a large spin disorder when combined with a large magnetic field. As a result, we have obtained a very large magnetoresistance effect in excess of 400 % at 4.2 K in a large magnetic field, the largest ever reported in any metallic systems. The STT characteristics of granular solids differ significantly from those of multilayers, showing no STT effect at low magnetic fields but prominent STT effects at high fields.

Giant magnetoresistance (GMR) [1] and spin-transfer torque (STT) effects [2-13] are two closely related phenomena of scientific and technological importance. Both GMR and STT effects were first observed in multilayers of ferromagnetic (F) and non-magnetic (N) metals, such as Co/Cu. The GMR effect is realized when a magnetic field alters the magnetic configuration of an F/N multilayer resulting in high and low resistances for antiparallel and parallel configurations of the F layers respectively. The STT effect is the inverse effect, where a spin-polarized current alters the magnetic configuration of the F layers. The STT effect is asymmetrical, causing parallel spin arrangements with one polarity of current and antiparallel configurations with the opposite polarity.

GMR occurs not only in multilayers, but also in granular solids, in which nanometer size magnetic entities (e.g., Co) are embedded in a non-magnetic medium (e.g., Cu or Ag) [14, 15]. At zero magnetic field, the randomly orientated magnetizations [15, 16] of the granules give rise to the high resistance state. When a magnetic field aligns the magnetic entities, GMR as high as 80 % has been observed [17].

In multilayers [4-11], a new phenomenon, precessing states with no time dependent driving, is induced by STT effects in large magnetic fields. The resistance of these states is in between that of the parallel and antiparallel states. In this work, we report the observation of yet another new form of STT that occurs in magnetic granular solids. In zero magnetic field, granular Co-Ag exhibits a GMR effect but no STT effect. At high fields and currents, we observe, in contrast, a new state which has a resistance much higher than those of the low-field states. The STT effect with a magnetic field generates a very large magnetoresistance (MR) in excess of 400 %, the largest GMR ever observed in any metallic systems, multilayers or granular solids.

We form granular $Co_{18}Ag_{82}$ (in volume percent) by exploiting the immiscibility between Co and Ag by co-sputtering of Co and Ag onto Si substrates at a Si temperature of $150^{\circ}C$ with a thickness ranging from 80 nm to 370 nm. As described elsewhere, we use non-magnetic metal point contacts (W, Ag, Cu) to accommodate the high current density necessary for the STT effects [4, 11-13]. Current flowing from the metal tip to the sample is taken as positive. The sample is housed in a vacuum vessel and placed in the liquid helium bath of a superconducting magnet system with the magnetic field mostly applied in the film plane (H), and occasionally perpendicular to the film plane (H_{\perp}), as schematically shown in inset of FIG. 1(a).

The resistance R = V/I of a granular Co₁₈Ag₈₂ film measured at 4.2 K using one point contact as a function of current I and magnetic field H is shown in FIG. 1(a) and 1(b) respectively. At a low current of I = 0.1 mA, GMR of 30 % is observed as expected (inset of FIG. 1(b)). When I is varied between +20 mA and -20 mA in H = 0, the value of $R \approx 4.6$ Ω is essentially unchanged as shown by the dashed curve in FIG. 1(a). The conspicuous absence of the STT effect in granular Co-Ag despite the presence of the GMR effect is distinctively different from the situation in multilayers, where both GMR and STT effects appear.

We have observed a dramatically different behavior under a large field of $\mu_0 H = 6$ T as shown by the red curves in FIG. 1(a). While R initially decreases to 3.34 Ω for positive I due to the normal GMR effect, R sharply increases at I = -4 mA, rising to 16.77 Ω . The spectacular increase in R is also revealed in the MR measurement as shown in FIG. 1(b). At I = 0.1 mA (blue curve) we observe the normal GMR of 30 %. However, at I = -7 mA (red

curve), R increases sharply from 3.34 Ω at 3 T to 16.77 Ω at 7 T. The five-fold increase in R, or an MR of 400 %, is by far the largest GMR ever reported in any metallic systems.

We have ruled out the possibility of mechanical artifacts by various measurements. At I = 4 mA, the current density through the contact has been estimated to be $7 \times 10^8 \text{A/cm}^2$, a value commonly encountered in point contacts and pillars. The 400 % MR is *not* due to electromigration or other high current density effects because it occurs only with one polarity of current *and* only under a large magnetic field. Instead, this effect is due to the special features of the STT effect in granular solids. In both the current scan (FIG. 1(a)) and the field scan (FIG. 1(b)), hysteretic reversal is observed, suggesting a magnetic origin of the unusual effects. Mechanical distortions can be ruled out because similar effects occur both with the magnetic field in the plane of the sample and perpendicular to it, as we discuss below. Further, these effects only occur for granular samples. We have not observed them in non-magnetic films, ferromagnetic films, or multilayers. Finally, effects of heating would not depend on the direction of the current. We conclude that the dramatic increase in resistance is due to a change in the magnetic configuration. We discuss a model for this behavior below.

At this point, it is useful to review the key aspects of the STT effects in multilayers, of which the simplest is a F1/N/F2 (e.g., Co/Cu/Co) trilayer consisting of a thicker F2 layer (the "polarizing" or "anchoring" layer) and a thinner F1 layer (the "free" layer) [3, 6-8]. When I exceeds I_C , the critical current for switching, the large angular momentum carried by the spin-polarized current switches the "free" F1 layer to have a parallel or antiparallel orientation relative to that of the F2 layer depending on the polarity of the current. The

resultant resistance difference induced by the STT effect is the same as that of the GMR. Switching by STT occurs at $H \neq 0$ as well, with I_C increasing linearly with H.

We have systematically measured the dependence of R on I and on H using another contact with $R \approx 2.4 \,\Omega$. As shown in FIG. 2(a), R is not appreciably affected by the current until the critical field of about $\mu_0 H_C = 4$ T, above which large increase in R occurs. As shown in FIG. 2(b), one observes only the normal GMR effect at low I within the field range of 9 T. Above the critical current $I_C \approx 16 \, \text{mA}$, R increases at high magnetic fields with a hysteretic behavior. We use the mid-points of the current-increasing branch and the field-increasing branch as representative values of I_C and I_C . It is clear that the larger the field the smaller the I_C . These values are plotted in FIG. 3(a) as the solid and open circles respectively. It may be noted that in trilayers STT switching exists at $I_C = 10 \, \text{m}$ with $I_C = 10 \, \text{m}$ with $I_C = 10 \, \text{m}$ with $I_C = 10 \, \text{m}$ as shown by the dashed straight line in FIG. 3(a). Exactly the opposite occurs in granular solids, where I_C decreases with increasing $I_C = 10 \, \text{m}$ to $I_C = 10 \, \text{m}$ as shown by the dashed straight line in FIG. 3(a).

Similar effects have been observed in over 40 contacts using 10 different samples. We note that with a low current, the measured resistance using the point-contact technique is $R_C + R(H)$, where R_C is the contact resistance and R(H) is mostly from the portion of the sample in the vicinity of the contact. The latter can be separately measured using the four-probe method and shows GMR of [R(0)-R(H)]/[R(H)] of 49 %. However, in the point-contact measurements, the apparent GMR of $[R(0)-R(H)]/[R_C+R(H)]$ is diluted by the sizable R_C , showing an apparent GMR between 10 % and 41 %. Those contacts (over 50 %) that have a higher GMR show the enhanced MR induced by the STT effect. The high field GMR ranges from 9 % to 400 %, with current densities between (3 to 9)×10⁸A/cm².

Experimentally, we have found that a large ordinary GMR (>20 %) is prerequisite for realizing the large MR induced by the STT effect with a large magnetic field.

As shown in FIG. 1 and 2, the STT effect seemingly occurs with only one polarity of H and I instead of both polarities. The asymmetry occurs on the opposite side by reversing either H or I. Closer examination of the results and more revealing differential measurements, show that there is indeed an accompanying feature, albeit much smaller, symmetrically located on the opposite side. We ascribe this acute apparent asymmetry to the Oersted field generated by the current through the structure, an effect that is frequently ignored [20]. Because of the smaller contact size than those of pillars, the Oersted field is larger at a similar current. As shown in the inset of FIG. 1(a), the Oersted field of the order of 1 T in the vicinity of the point contact is added to, or subtracted from, the applied magnetic field depending on the direction of the in-plane H and the locations of the electrical contacts on the sample. Indeed, as demonstrated in FIG. 2(c), under a perpendicular H_{\perp} , the STT effect occurs at both polarities. This result also shows that the observed spectacular MR is not due to any force on the current-carrying contact by the magnetic field.

The resistance increase induced by the STT effect in conjunction with a magnetic field becomes much smaller at a higher temperature of 160 K. This is because some of the small Co granules, switched through the STT effect at low temperature, are now superparamagnetic, thus compromising the antiferromagnetic spin structure. However, the STT events can still be observed in more sensitive differential resistance (dV/dI) measurements as shown in FIG. 4(a) covering both polarities of I and H. The "ridge" structure at $H \approx 0$ is the normal GMR effect, but all the features at higher H are due to the

STT effect. The four branches of loci are at the onset of reversal, whose values display the expected symmetry with respect to *H* and to *I* as shown in FIG. 4(b). Comparing with those at 4.2 K, similar currents and fields are required at 160 K to realize the STT effects of smaller magnitude in resistance. The strong temperature dependence also attests that the observed STT effect is not a mechanical artifact.

The following scenario explains our observation of resistance four times larger than that of the parallel state and also much larger than the disordered remnant state. In the remnant state, the magnetizations have random orientations with no net magnetization as indicated by the schematic spin structure near point a in FIG. 1(b). As a magnetic field is applied, the magnetizations are pulled into a low resistance parallel configuration as indicated by the schematic spin structure near point b in FIG. 1(b). The resistance of this configuration is lower because a uniform spin polarized current allows most of the current to flow through the low resistance majority channel. We believe the high resistance state reached by applying a large current is a state with collinear moments but reduced magnetization because a number of the moments are antiparallel to the others as indicated by the schematic spin structure near point c in FIG. 1(b). This state has the highest resistance because the spin diffusion length is much longer than the intergranular separation, leading to no current polarization. The resistance of the remnant configuration is much lower because non-collinear magnetizations act as strong spin-flip scatterers leading to local, although not global, polarization of the current.

The collinear state with reduced magnetization is stabilized only in the presence of a large field and a large current density for the following reasons. In granular Co-Ag solids, the Co granules are similar but different in sizes. A large *H* aligns the magnetizations in a

torque free state. An antiparallel configuration of moments is also torque free, but unstable in the absence of current. However, a large polarized current can stabilize this configuration. The largest Co granules, serving the anchoring role, remain aligned with H as I is increased. Increasing the current from small values causes the magnetization of some grains to reverse, increasing the resistance even at small magnetic fields and small currents as shown in FIG. 3(b), where R at 6 mA and 10 mA is significantly larger than that at 0.1 mA. The reduction of the total moment reduces the spin polarization of the current, giving rise to a higher resistance. At a higher H, the STT effect causes an even higher degree of spin misalignment and eventually compels the reversal of some of the granules resulting in even higher resistances. As I is further increased, other intermediate size Co granules follow suit in a cascading manner until a maximum degree of antiferromagnetic alignment of the Co magnetic moments (denoted as c in FIG. 1(b)) is achieved with the largest resistance. At this point, the STT effect at a given H is self-limiting. During this reversal process, some smaller Co granules may reverse their magnetizations more than once in order to achieve the highest spin disorder. While the states a and b of the normal GMR are thermodynamically stable, the state c, achieved under a large current and a large field, is metastable. This fact is manifested by the much higher noise of state c than those of either a or b as shown in FIG. 1.

Typical values of GMR in multiayers are around 10 %, but GMR of 150 % has been observed in epitaxial Fe/Cr multilayers [18], and even larger values were found in calculations [19]. Granular solids with nanometer size magnetic granules have a higher density of interfaces which can potentially lead to a larger GMR. However, the random spin structure with zero magnetization, the high resistance state of the normal GMR, is not

the one with the largest resistance. In granular solids, the antiferromagnetic spin structure holds the largest resistance but is not reachable by the application of a magnetic field. Through the STT effect with a large magnetic field, we have found the elusive antiferromagnetic spin structure in granular solids, and captured a spectacular MR of 400 % in a system with a normal GMR of only 30 %.

Finally, as shown in FIG. 3(a), the $|I_C|$ of multilayers and granular solids has the opposite dependence on $|H_C|$ because the mechanisms are different. The magnetic field does not affect the "anchoring" layer but only impedes the reversal of the "free" layer, thus the value of $|I_C|$ increases linearly with $|H_C|$. In granular solids, in the absence of H, there is no net magnetization or spin polarization. The spin polarization depends on the degree of alignment of the nm-sized Co granules by the H field. As H increases, the STT effect becomes stronger due to the larger spin polarization, hence the value of $|I_C|$ decreases with $|H_C|$, exactly opposite to that in multilayers.

In conclusion, we have demonstrated a new form of STT effect in conjunction with a magnetic field in a granular Co-Ag solid. The combined effect of a large current and a large magnetic field creates a maximum spin disorder resulting in a record GMR in excess of 400%. The characteristics of the STT effect in granular solids are much different than those previously observed in multilayers.

Work supported by NSF Grant Nos. DMR04-03849 and 05-20491.

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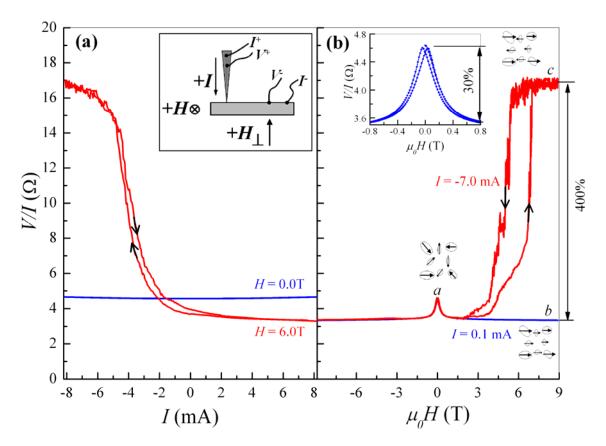


FIG. 1. (a) Resistance versus current in 0 T (blue) and 6 T (red) of a contact on granular $Co_{18}Ag_{82}$ (sample #12) at 4.2 K, inset shows schematics of point contact measurement, (b) magnetoresistance (MR) of 30 % (blue) using currents 0.1 mA and 400 % (red) MR at -7 mA of the same contact as in (a), and the schematic spin structures at a, b, and c; the inset shows MR in small fields.

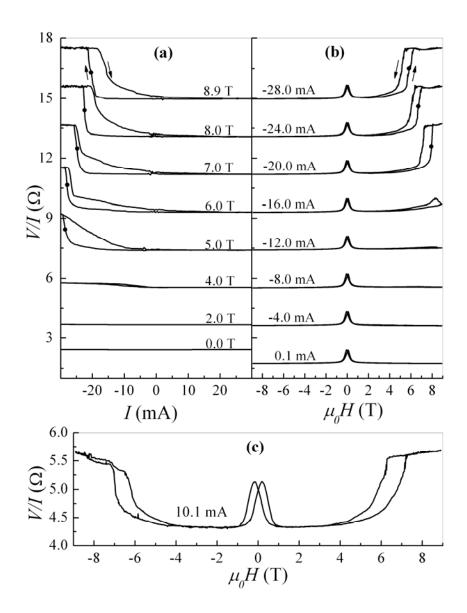


FIG. 2. (a) Resistance V/I versus current I at different fields H in 1 T interval and (b) Resistance vs. H at different I in 4 mA interval using one point-contact on $Co_{18}Ag_{82}$ (sample #12) at 4.2 K. (c) R vs. H at 10.1 mA using another contact with H perpendicular to the film plane on $Co_{18}Ag_{82}$ (sample #14) at 4.2 K.

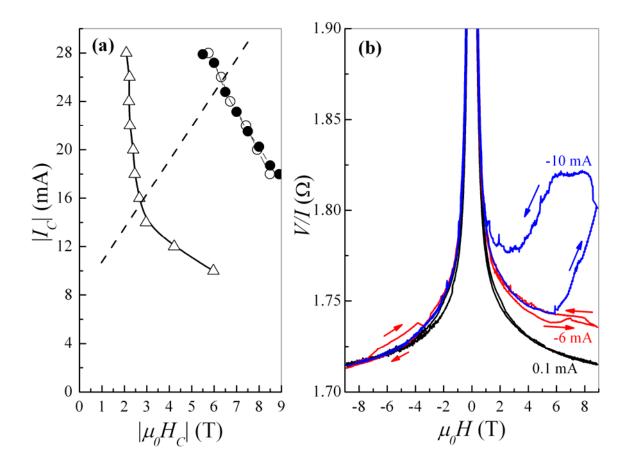


FIG. 3: (a) Dependence of critical switching current I_C on critical field H_C in Co₁₈Ag₈₂: open and closed circles are the mid-point values for the increasing H and I branch respectively in FIG. 2(a) and 2(b), and triangles are the locations where R begins to increase. The dashed line shows the corresponding dependence in multilayers [7], (b) MR results at I = 0.1 mA (black), -6 mA (red) and -10 mA (blue) of the same contact in FIG. (a) and 2(b).

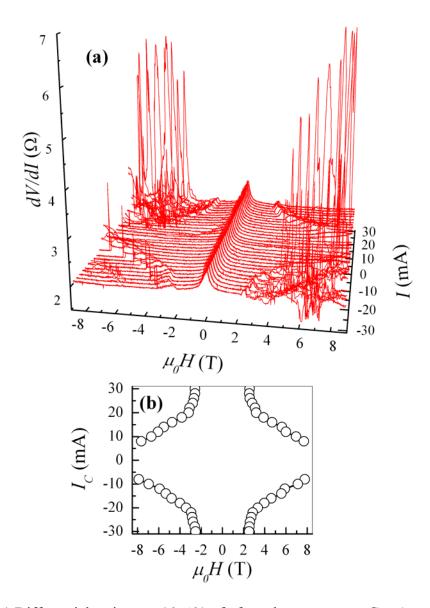


FIG. 4: (a) Differential resistance (dV/dI) of of another contact on Co₁₈Ag₈₂ (sample #12) at 160 K at various currents and magnetic fields, (b) four branches of loci at the onset of switching.